FIRST-Nuclides: Outcome, Open Questions and Steps Forward

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**Associated Groups**

13 Groups participating at their own costs with specific RTD contributions or particular information exchange functions, or mobility measures.

**End User Group**

Review the scientific-technical progress and contributions to the proceedings and assess the project status and provide recommendations for the work program.
Objectives

- Experimental
  - Quantification of relation FGR to IRF for $^{129}$I, $^{79}$Se, $^{135}$Cs
    - for relevant burn-up / lin. power rate ranges,
    - for full set of sample sizes (pellet, ..., powder),
    - for same groundwater different atmospheric conditions
  - If possible: Quantification (speciation) of elements such as $^{14}$C, Se

- Modelling
  - Up-scaling

- Training, Education, Dissemination

- Further expected outcome of FIRST-Nuclides
  - Definition of instant – rapid – fast release and delineation to long-term RN release processes.
  - Up-scaling from lab sample to rod/assembly.
  - Effect of geochemical conditions.
Materials

Selection, characterization and preparation of materials and set-up of tools

- Characterization of selected SNF materials with respect to
  - Fuel characteristics, irradiation history,
  - Permission by the fuel owner for publication of key parameters
- Preparation of SNF samples, cladded pellets, pellets, powders, etc.
- Experimental set-ups (autoclaves, irradiation cells, reaction vessels),
  - Sampling devices
  - Analytical equipment in the hot cells

WP leader: Volker Metz (KIT-INE)
### Selected high burn-up $\text{UO}_2$ fuel samples

**Relevant SNF samples**

<table>
<thead>
<tr>
<th>Units</th>
<th>PWR</th>
<th>BWR</th>
<th>THTR / VVER</th>
</tr>
</thead>
<tbody>
<tr>
<td>Enrichment</td>
<td>%</td>
<td>3.80 – 4.94 %</td>
<td>3.30 - 4.25 %</td>
</tr>
<tr>
<td>Burn-up</td>
<td>GWd/t$_{\text{HM}}$</td>
<td>50.4 – 70.2</td>
<td>48.3 – 57.5</td>
</tr>
<tr>
<td>lin. power</td>
<td>W/cm</td>
<td>186 - 330</td>
<td>160</td>
</tr>
<tr>
<td>FGR</td>
<td>%</td>
<td>4.9 – 23 %</td>
<td>1.2 – 3.1 %</td>
</tr>
</tbody>
</table>
**WP 2: Gas release and rim and grain boundary diffusion**

Direct comparison between FGR and IRF

Radial FP distribution over the pellet radius by laser ablation mass spectroscopy

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**WP 2: Non LWR materials**

IRF from spent UO2 TRISO coated particles, microstructure evolution

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WP leader: Detlef Wegen (JRC-ITU)
Results on speciation of Selenium

XANES measurements: mixture Se(0) and Se(IV) or pure Se(-II)
Modelling: Homogeneous chemical form, probably as Se(-II) replacing oxygen sites in the UO₂ lattice.
Cooperation PSI & Studsvik

Behaviour of $^{14}$C

Dissolution based measurements:

$\Rightarrow IRF(^{14}\text{C}):$ below 1.5% ✓

$^{14}$C in the plenum gas measurement:

$\Rightarrow IRF(^{14}\text{CO}_2 \text{ gas}) = 0.2 \% \text{ of } ^{14}\text{C inventory}$ ✓
**WP 3: Dissolution based release**

- Dissolution based radionuclide release and to the extent possible the chemical speciation of the relevant isotopes.

### Leachants

<table>
<thead>
<tr>
<th></th>
<th>NaCl</th>
<th>NaHCO$_3$</th>
<th>pH</th>
<th>Conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>PSI</td>
<td>19</td>
<td>1</td>
<td>8.5</td>
<td>anoxic / slightly oxidizing</td>
</tr>
<tr>
<td>KIT</td>
<td>19</td>
<td>1</td>
<td>8.9</td>
<td>Ar/H$_2$ (Eh = -116 mV)</td>
</tr>
<tr>
<td>ITU/CTM</td>
<td>19</td>
<td>1</td>
<td>7.4</td>
<td>Oxidizing (air)</td>
</tr>
<tr>
<td>SCK-CEN</td>
<td>19</td>
<td>1</td>
<td></td>
<td>anoxic / slightly oxidizing</td>
</tr>
<tr>
<td>Studvik</td>
<td>10</td>
<td>2</td>
<td>8.1-8.2</td>
<td>Oxidizing (air)</td>
</tr>
</tbody>
</table>

WP leader: Karel Lemmens (SCK-CEN)
IRF_{Cs} of different fuel / sample sizes
IRF Cs: Delineation from long-term release

- Powder
- Single fragments
- Pellets
- Slices of pellets

FIAPCs/day (long term experiments)

Matrix

Gap

Grain boundaries

Log fractional release rate (days⁻¹)

Log Time / days
IRF$_{\text{iodine}}$ of different fuel / sample sizes

![Graph showing IRF$_{\text{iodine}}$ over time for different samples]
IRF as function of burn-up / linear power

IRF: no correlation with burn-up

Pellets

Uncladded, fragments,

IRF: 150-180 days
Effect of reducing conditions on IRF Rates (Sr, U)

![Graph showing the effect of reducing conditions on IRF Rates (Sr, U)]
Dissemination

1st Annual Workshop Proceedings of the Collaborative Project 'FIRST-Nuclides'
KIT SR 7639 (2013) Download: http://dx.doi.org/10.5445/KSP/1000032486

2nd Annual Workshop Proceedings of the Collaborative Project 'FIRST-Nuclides'
KIT SR 7676 (2014)

Training Courses:
Topical sessions during the AWS Young Scientists Training Course, July 09 – 10, 2013
Young Scientists Mobility Measures

Involvement of non-scientific stakeholders:
Meeting with InSOTEC representatives, July 2013
Presentation of InSOTEC outcome @ 2nd AWS

27th Spent Fuel Workshop:
9 oral talks of FIRST-Nuclides results

WP leader: Alba Valls (Amphos21)
Findings

- Release depends on sample characteristics:
  - on the operation parameters, burnup and power rate
  - on the nature of the exposed fuel structures (gap or grain boundaries)
  - Release tends to increase in the order: fragments < clad pellet segments < opened clad pellets, (exposed surface area / gap inventory)
  - Al/Cr doped fuel have lower IRF (larger grain size)
  - IRF higher for PWR fuel as for BWR fuel (linear power)

- Effect of redox conditions for U, Sr, Tc
  - Significant differences for U, Sr and Tc under oxidizing or reducing conditions

- IRF$_{Se/14C}$: few data available presently. Measurements not yet completed.

Evaluation of data obtained in FIRST-Nuclides not yet completed.
End-User Comments

- FIRST-Nuclide is highly relevant for all WMO
- IRF contributes substantially to the peak releases after container breaching and to potential radiological consequences.

Results include:

- Data from experimental determination of rapid release fractions for moderate and high burn-up UO₂ fuels.
- Doped fuels, expected to be used much more in the future.
- Increased data base for release of Cs and I from high burn-up fuel
- Comprehensive comparisons of IRF with fission gas release (FGR)
- Basis to estimate IRF data for a very large number of fuel rods and various reactor operation conditions.
- Improvement of analytical techniques
Conclusions

- Successful project
- Improved understanding of “IRF”
- Correlations between “reactor data” and IRF
- Publications in preparation
- Huge investments on
  - setting-up the experiments,
  - implementing the required analytical tools and instruments and
  - clearance by the utilities to publish the spent fuel data.
- Experimental duration could be extended
- Open issues still exist
Steps Forward

- Continuation of the 3 years project
  - maximize the outcome
  - maximal exploitation of investments (hot cell installations, analytics, staff, ...)
- Improved statistics for the IRF of other fission products.
- In depth investigations of low concentrated but relevant isotopes $^{79}\text{Se}$, and $^{107}\text{Pd}$, or activation products $^{36}\text{Cl}$ and $^{14}\text{C}$.
- Experiments under repository relevant conditions, i.e. reducing conditions, H2 overpressure
- Correlations for predicting the IRF from nuclear power plant operational parameters (power rates, ramps, temperatures, FGR).
- Delineating the instant release from long-term radionuclide release.
- Additional kind of samples (e.g. MOX, reprocessed uranium fuel, low-FGR fuels, ...)
- Keeping the know-how.
Proposed Procedure

- Keeping the experimental set-up and the materials for a certain interim period in the labs
- Depending on financial support by the WMO
  - Less frequent sampling
  - Storage of samples
- Required support by the WMO/IGD-TP:
  - New project at the next EU Call
  - Materials from utilities (transports, etc.)
  - Receiving operational parameters from the utilities such as power rates, ramps, temperatures, FGR
    - Not only average values!